

# **NUCLEAR ENERGY RESEARCH INITIATIVE**

## **6. NUCLEAR WASTE MANAGEMENT**

This program area addresses the long-term R&D goal related to fuel cycle research which considers the impact of fuel cycle options on waste generation, waste form, and waste storage and disposal.

Projects currently funded include R&D that address nuclear waste technological improvements to the back end fuel cycle process. Novel approaches are proposed to reduce the physical volume of spent nuclear fuel and to recycle or reuse spent nuclear fuel without reprocessing in a manner that maintains the highest degree of proliferation resistance. Additional R&D is being performed in the use of concrete in nuclear waste containment.

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## **Monitoring the Durability Performance of Concrete in Nuclear Waste Containment**

PI: Franz-Josef Ulm, Massachusetts Institute of Technology

Collaborators: Commissariat à l'Energie Atomique (French Atomic Energy Commission)

Project Start Date: August 1, 1999                      Projected End Date: August 31, 2002

Project Number: 99-0126

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### **Research Objective**

Concrete is commonly employed in radioactive waste disposal as an effective construction material for containment barriers, liners, and encasement of containers. The objective of this research is to develop the scientific knowledge and the appropriate engineering tools required to evaluate and quantify the durability performance of nuclear waste concrete containment subjected to the pessimistic chemical degradation scenario of calcium leaching. Monitoring the durability performance means here the quantitative assessment, in time and space, of the integrity of the containment during the entire storage period and requires the consideration of the multiple couplings between diffusion-dissolution of calcium and deformation and cracking.

With regard to the time-scale, the durability design of waste containers needs to consider some reference scenario of chemical degradation, in particular the pessimistic one of calcium leaching by pure water. This design scenario refers to the risk of water intrusion in the storage system. For the reference scenario at hand, it is generally assumed that concrete is subject to leaching by permanently renewed deionized water acting as a solvent. The calcium ion concentration in the interstitial pore solution leads to dissolution of the calcium bound in the skeleton of Portlandite Crystals,  $\text{Ca}(\text{OH})_2$ , and calcium-silica-hydrates (C-S-H), with sharp dissolution fronts. This calcium leaching leads to a degradation of the mechanical properties of concrete (material strength, Young's modulus). Cracks increase the diffusivity of the calcium ions through the structure, and can lead to an acceleration of the chemical degradation, and hence to an acceleration of the overall structural aging kinetics. This process can lead to a closed loop of accelerated structural degradation.

### **Research Progress**

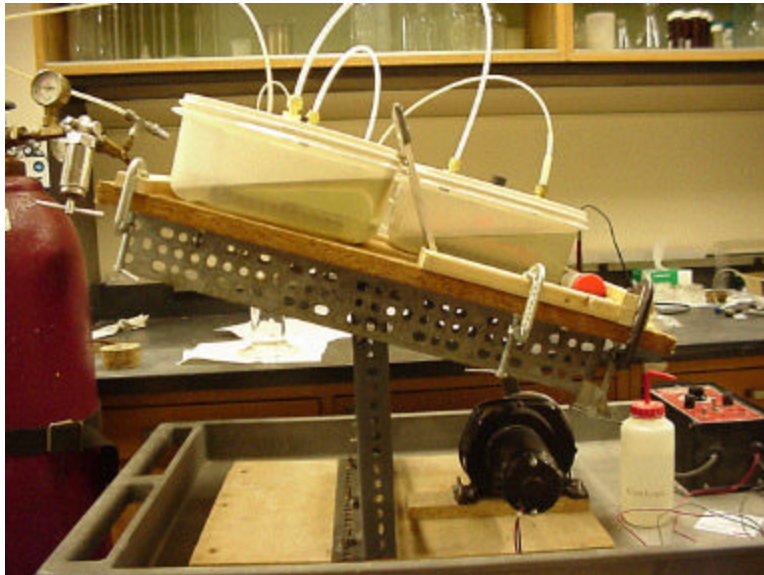
After the first year of research, the important scientific findings that will translate into industrial benefits in the field of concrete durability in nuclear waste storage are as follows:

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- The chemical process of calcium leaching involving kinetics and mineral composition were studied, including the use of alternative leaching agents. State-of-the-art material tests were employed.
- The consequences of calcium leaching on the mechanical behavior of cement paste were identified in mechanical tests, invoking three dimensional (3-D) stress states. For the first time, the governing mechanical parameters were identified in 3-D.
- The role of cracks on demineralization of porous materials was the object of an extensive study. By means of dimensional analysis, the governing similarity properties of calcium leaching and degradation of concrete were identified in an unprecedented completeness.
- In particular, making use of similarity properties of the governing equations of 'real' calcium leaching in concrete, it was shown that a pure diffusive mass transport through a crack or fracture will not significantly affect the overall degradation kinetics of a concrete structure, while advective transport may accelerate the degradation process.

The resulting industrial benefits can be summarized as follows:

- Based on the scientific analysis of the calcium leaching process, an accelerated material leaching test was conceived and put into practice, allowing for 300-fold accelerated calcium leaching. The developed test setup is illustrated in the picture below. This makes it possible to test different kinds of cementitious materials with regard to their leaching characteristics before being utilized in industrial applications.
- Changes in mineral composition due to calcium leaching are now predictable and can be implemented in industrial planning and design tools. For instance, the thickness of

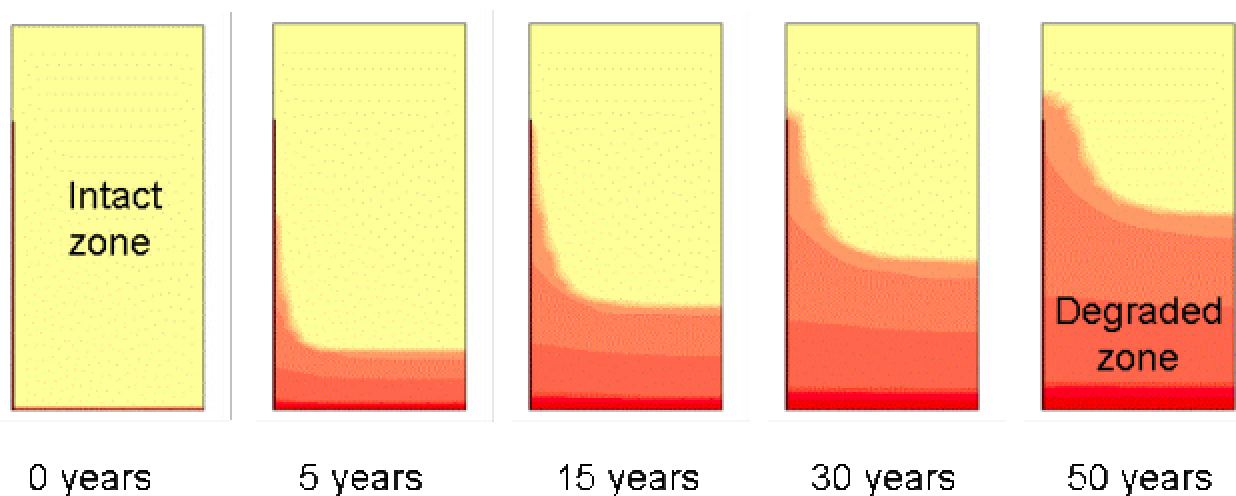


**Experimental setup for accelerated calcium leaching of cement-based materials:** Material samples are immersed in a 6M ammonium nitrate solution for accelerated leaching. In order to obtain good mixing of the ammonium nitrate solution and most homogeneous leaching conditions possible, tanks containing the ammonium nitrate bath are mounted on a slowly oscillating table.

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containment structures can be adopted with regard to the demineralization design scenario.

- The results of mechanical testing are a key element for the mechanical modeling of calcium leaching, needed for industrial monitoring and decision support systems for safe storage systems.
- The evolution of a calcium-leaching front was implemented successfully in a commercial finite element code. This is a necessary condition for transfer of the research results into engineering practice. The figure below shows a typical example of the propagation of a dissolution front around a fracture that can be obtained by means of the developed program. This model can be readily employed for long-term structural leaching analysis.
- The profound analysis of influential parameters concerning cracks in cementitious materials and their consequences on leaching have immediate industrial application. For example, it makes it possible to analyze the suitability for industrial applications of a given cementitious material or structure at any given moment during its life span. This can improve the decision quality compared with a decision that solely considers the crack size, as is common practice in parts of industry today.
- The issue of nuclear waste storage and the corresponding initiatives of the U.S. government through DOE's NERI program are brought to a large audience consisting of both the scientific-engineering community and the broader public.



**Propagation of the dissolution front around a crack (left of the figure) obtained by model-based simulations. This type of results indicate that small fractures do not significantly increase the overall deterioration kinetics of calcium leaching.**

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## **Planned Activities**

The next steps of the project will include the completion of the experimental studies on the material and structural level. In addition, the computational mechanics approach will be extended, developing the mechanical model accounting for the chemo-hygral-mechanical cross effects. Focus of this combined experimental-theoretical approach will be on bridging scales from the microscale of the experimentally assessed material deterioration to the macroscale of engineering prediction of the performance of concrete employed in nuclear waste repositories.

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## Chemical Speciation of Neptunium in Spent Fuel

PI: Ken R. Czerwinski, Massachusetts Institute of Technology

Collaborators: Argonne National Laboratory

Project Start Date: August 1, 1999      Projected End Date: August 31, 2002

Project Number: 99-0127

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### Research Objective

This project will examine the chemical speciation of neptunium in spent nuclear fuel. Neptunium has been identified as a radionuclide of concern and information on the chemical form of neptunium in spent fuel is lacking. The identification of the neptunium species in spent fuel would allow a greater scientific based understanding of its long-term fate and behavior in waste forms.

The chemical species and oxidation state of neptunium will be determined in spent fuel and alteration phases. Different types of spent fuel will be examined. Once characterized, the chemical behavior of the identified neptunium species will be evaluated if it is not present in the literature. Special attention will be given to the behavior of the neptunium species under typical repository near-field conditions (elevated temperature, high pH, varying Eh). This will permit a timely inclusion of project results into near-field geochemical models. Additionally, project results and methodologies have applications to neptunium in the environment, or treatment of neptunium containing waste.

### Research Progress

Speciation of Neptunium in Spent Fuel and Alteration Products: The application of synchrotron-based methods, X-ray absorption near edge spectroscopy (XANES) and extended X-ray absorption fine structure (EXAFS), to the identification and characterization of environmentally relevant solids is well established. The application of synchrotron methods to establish the oxidation states of actinides in precipitated, altered, and unaltered neptunium phases has also been demonstrated. Progress continues with the characterization of carefully prepared neptunium standards to build a library of spectra that will help establish the nature of the unknowns. Preparations for synchrotron studies of neptunium-bearing solids were also completed.

Analyze Np Solid Phases and Information of Np Alteration Phases: Progress in these areas involved examining two separate efforts; the subsurface chemistry of neptunium when organics are present and the effects of microbiological activity on neptunium speciation and phase alteration. The reactivity of Np(VI) with citrate was investigated as

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a model system to understand the role of complexation and redox conditions in defining the oxidation state distribution under subsurface conditions. It was established that citrate reduces Np(VI) to Np(V) within days under all conditions investigated. The disappearance of Np(VI) was first-order under all conditions investigated but the pseudo first order rate constant was not proportional to the concentration of citrate due to changes in the reaction mechanism.

Research was also initiated to establish the effects of microbiological activity on the speciation of neptunium. Emphasis is on a *Shewanella* strain that is an anaerobic metal-reducing bacterium common to many subsurface aquifers. The methodology for growing and studying this isolate was established. Interaction experiments with neptunium are planned for the early part of the second year in the project.

Behavior of Np Species with Ligands: The sorption and precipitation of Np(V) on goethite, montmorillonite, and tuff was studied. The first experiment was done in 0.1 M NaClO<sub>4</sub>, at room temperature, and in a nitrogen glove box. The second experiment was done in 0.1M NaClO<sub>4</sub>, at room temperature, and under an air atmosphere. The third experiment was done in J-13 groundwater, at room temperature, and under an air atmosphere. Supernate concentrations were measured and the solid phase was analyzed at the Advanced Photon Source at ANL. Current results show sorption of Np(V) to goethite, montmorillonite, and tuff. The proton exchange capacity was found to be 0.228±0.018 meq OH/g for FeOOH and 0.770±0.016 meq OH/g for montmorillonite. This data will be used to model Np(V) sorption.

In further studies under inert atmosphere the supernate concentrations in the goethite samples dropped to approximately 60 percent of the original Np concentration at low pH, and decreased to very low concentrations by pH 7.5. The Np concentrations in the tuff samples were slightly higher than those in the goethite samples at low pH. This trend continued until approximately pH 9.5, which is the pH where hydrolysis is predicted to begin. At this point, Np concentration decreased dramatically until pH 11. After pH 11, Np concentration increased again. This suggests a pH dependent precipitation of Np within the tuff system or a pH dependent sorption effect. Previous studies elsewhere found sorption within the pH range of 6.5 to 8.5, but also found that some tuff samples exhibited negligible sorption. Further studies are necessary to better understand the change in supernate concentration as a function of pH.

Data for Modeling: Modeling of Np under Yucca Mountain conditions from pH 5 to pH 12, Eh from 0 V to 1.5 V and temperature from 25 °C to 100 °C was performed with CHESS 2.4. Seven different main Np species were observed in the calculations. In conditions below 0.5 V the tetravalent NpO<sub>2</sub> is formed. Three pentavalent Np species, NpO<sub>2</sub><sup>+</sup>, NpO<sub>2</sub>CO<sub>3</sub><sup>-</sup>, NpO<sub>2</sub>OH<sub>(aq)</sub> are dominant in the potential ranging from 0.5 V to 1.15 V. Three hexavalent Np species: NpO<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub><sup>4+</sup>, NpO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub><sup>2+</sup>, NpO<sub>2</sub>OH<sup>+</sup>, are in the oxidizing conditions above 1.15 V. Other species with fluoride, sulfate and phosphate are calculated to exist but at very low concentrations. The database lack mixed ligand species or possible Np mineral type phases that may be found in spent fuel. In addition,



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the predicted behavior of  $\text{NpO}_2$  indicates future efforts need to examine the dissolution kinetics.

Electrodeposition system for alpha spectroscopy: Electrodeposition has been shown to be the most effective method of actinide sample preparation for alpha spectroscopy. An electrodeposition apparatus was made in order to meet the laboratory alpha spectroscopy sample preparation needs associated with the Np speciation project.

## **Planned Activities**

- Develop methods and equipment for performing XANES/EXAFS experiments with spent fuel.
- Systematically analyze and characterize a wide range of oxides, hydroxides, carbonates, phosphates, silicates and environmental complexes to identify the atomic and structural contributions to the XANES/EXAFS and other solid phase analysis.
- Calculate the key structures of Np species.
- Analyze Np and alteration products.
- Determine the solubility and chemical behavior of Np species examined in earlier studies. Evaluate the effect of temperature and environmental ligands on Np speciation.
- Use project data to evaluate the behavior of Np in the repository.

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## Experimental Investigation of Burn-up Credit for Safe Transport, Storage, and Disposal of Spent Nuclear Fuel

PI: Gary A. Harms, Sandia National Laboratories

Project Start Date: August 1, 1999

Projected End Date: August 31, 2002

Project Number: 99-0200

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### Research Objective

The Nuclear Energy Research Initiative has funded a critical experiment focused on burnup credit issues at Sandia National Laboratories. The experiment, when complete, will provide benchmark data that can be used to test the methods and data used in the criticality safety analyses of shipping, storage, and disposal of spent nuclear fuel.

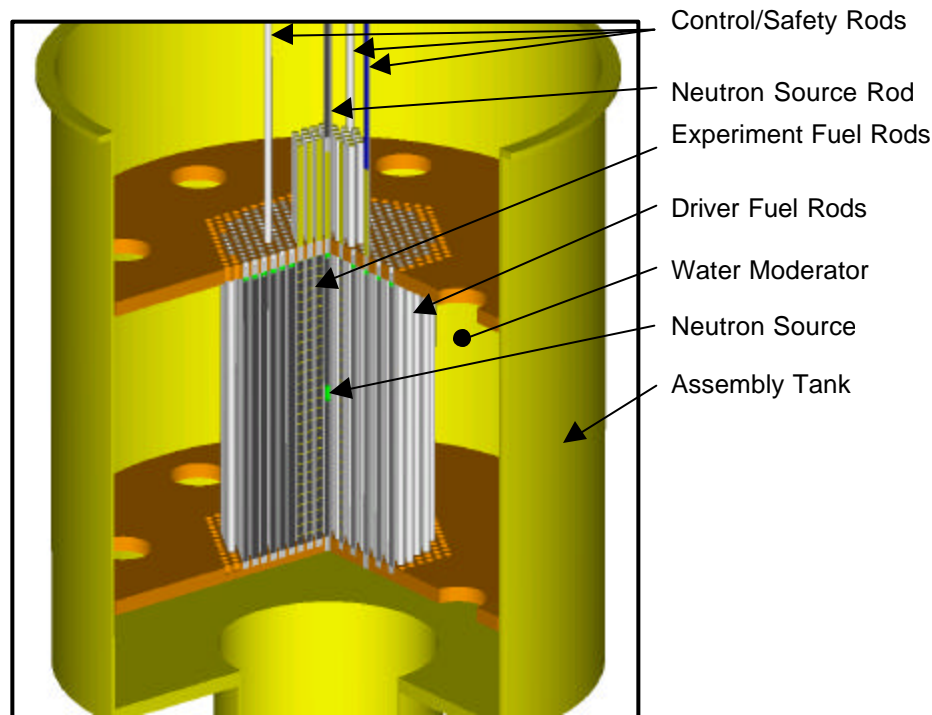
Burnup credit is the process of accounting for the decrease in the reactivity of spent nuclear fuel produced by the changes in the fuel actinide concentrations and the buildup of fission product absorbers caused by the burning of the fuel. To apply burnup credit safely, the methods used in its application must be validated: the fuel isotopic composition in the burned state must be accurately predicted, and the neutron multiplication of spent fuel configurations must be accurately predicted. The current experiment addresses the second part of the validation issue.

A critical assembly with low-enriched  $\text{UO}_2$  fuel will be built at Sandia. The assembly concept is shown in the figure below. It consists of a water-moderated array of driver fuel rods surrounding a smaller number of experiment fuel rods. The experiment fuel rods will allow for the insertion of test materials between the fuel pellets in these special fuel rods. The assembly also includes three fuel-followed control/safety rods and a fueled source rod. Critical experiments will be performed in the unperturbed assembly and with a test material, a fission product simulant, present in the assembly. The benchmark data from the experiment will be the difference in the critical array size between the two configurations.

The critical experiment is structured as a three-year project. The first two years will be used to obtain the necessary approvals for a new nuclear experiment and to design and procure the experiment hardware. The benchmark experiments will be performed in the third year. Five broad tasks were outlined in the project proposal:

- Task 1 - Obtain the necessary National Environmental Policy Act (NEPA) approvals.
- Task 2 - Prepare safety basis documentation for the experiment and obtain approvals.
- Task 3 - Design and procure the experiment hardware.
- Task 4 - Perform the benchmark experiments.
- Task 5 - Decommission and decontaminate the experiment.

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**Layout of the critical assembly**

## **Research Progress**

In the first year of the project, work was performed on Tasks 1 through 3. The NEPA approvals for the experiment were obtained. Sandia filed an Environmental Checklist/Action Description Memorandum with the Department of Energy requesting that the experiment be categorically excluded from further NEPA documentation. The exclusion was granted.

The preparation of the safety basis for the experiment was started and will continue into the second year of the project. An Unreviewed Safety Question Determination (USQD) for the experiment was prepared against the safety bases of the Sandia Pulsed Reactor facility. Since the experiment differs significantly from the bare metal reactors normally operated in the facility, it was felt that the Technical Safety Requirements did not adequately bound the operation of the experiment. As a result, the USQD was positive. An addendum to the facility Safety Analysis Report is being prepared. A draft of the addendum is complete and under internal project review. Review by the Sandia Internal Review and Appraisal System and by the Department of Energy is yet to be accomplished.

The design and procurement of the experiment hardware was started and will continue into the second year of the project. The design of the critical assembly (the hardware in which the experiment is performed) is based on an earlier critical assembly that was built

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and operated at Sandia. Fuel for the current experiment was built for another Sandia burnup credit critical experiment that was never conducted. The hardware and fuel from the earlier experiments were archived temporarily at Los Alamos National Laboratory. This past year, both the hardware and the fuel were returned to Sandia. The design of additional hardware specific to the current assembly was started and continues.

Conceptual designs for the critical experiments were started. As part of the design process, an *ad hoc* panel of burnup credit experts was formed and asked to provide programmatic input to the design of the critical experiments. With this input, a conceptual design of the first experiment was completed. Experiment design will continue throughout the project.

## **Planned Activities**

Final approval of the safety bases for the experiment will be sought in the second year of the project.

The design and procurement of the assembly hardware will continue into and be completed in the second year of the project. The assembly is expected to commence operation at the beginning of the third year of the project.

The benchmark critical experiments will be run in the third year of the project. Design activities for the experiments will continue in the second year and be complete when the experiments are finished. The experiments will be documented upon completion.

After the experiments are complete, and if no further experiments are planned, the assembly will be decontaminated and decommissioned in the third year of the project.

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## **Deterministic Prediction of Corrosion Damage in High Level Nuclear Waste**

PI: George Engelhardt, SRI International

Collaborators: Pennsylvania State University

Project Start Date: August 1, 1999                      Projected End Date: August 31, 2002

Project Number: 99-0217

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### **Research Objective**

This research involves developing deterministic models and associated computer codes for predicting the evolution of corrosion damage to high level nuclear waste (HLNW) containers. Safe disposal of our nation's HLNW represents one of the greatest technical challenges of the twentieth and twenty-first centuries. The principal challenge is to ensure isolation of the waste from the biosphere for periods up to 10,000 years under conditions that can only now be estimated. The lack of existing databases for the corrosion of candidate alloys over times that represent even a small fraction of the intended service life means that we cannot rely on empirical methods to provide the design, materials selection, and reliability assessment information that is required to assure the public that the technology chosen for the disposal of HLNW is effective and safe. Instead, only strategies based on the employment of deterministic models can be used, because the natural laws (laws of conservation) that are the foundation of these models constrain the solutions to physical reality and are invariant with time.

Existing deterministic models of general and localized corrosion allow us to predict the accumulation of corrosion damage in many systems. However, these models must be customized for predicting damage in HLNW canisters in a tuff repository. Thus, the influence of radiolysis on the corrosion potential and hence on the corrosion rate, for example, must be included in the models. Particular attention must be given to repassivation phenomena, because they eventually determine the extent of damage. Attempts to quantitatively describe localized corrosion damage without proper consideration of repassivation phenomena greatly underestimate the service lives of containers. It is also important to customize the models to the conditions to which the containers are expected to be exposed over their design lives.

The principal objectives of this project are to:

- Develop deterministic models and associated computer codes for predicting the evolution of corrosion damage (i.e., 'integrate' damage) to HLNW containers in the Yucca Mountain repository. Corrosion processes that will be considered include

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general corrosion (oxidation), pitting corrosion, crevice corrosion, and stress corrosion cracking.

- Develop deterministic methods for extrapolating corrosion rate data obtained under “accelerated” laboratory conditions, to the field.
- Use the models to predict the fates of containers after exposure in the repository under various conditions (e.g., humid air, contact with dripping water, repository inundation).

It is evident that the first objective is the basis of the project, and that other objectives can be achieved only if deterministic models for predicting corrosion damage to HLNW containers are developed.

## Research Progress

The following accomplishments have been achieved during the first year of research:

- The differential equation for calculating the damage function (DF) for localized corrosion has been derived. By analytical or numerical solution of this equation, it is possible to calculate DF under arbitrary conditions if the rate of nucleation and propagation and the probability of survival of corrosion events are known.
- A general computer code for calculating potential and concentration distributions in corrosion cavities has been developed.
- Simple, approximate analytical (but rather accurate) expressions for calculating potential and concentration distributions along with the cavity propagation rates for Alloy C-22 have been developed. This development is very important to the ultimate success of this program, because accurate numerical simulation of corrosion damage for a long period (up to 10,000 years) may require excessive amounts of computer time.
- Detailed radiochemical simulations of the effects of ionizing gamma and neutron radiation on the properties and chemical composition of electrolyte solutions under repository conditions indicate that the impact on pH should be minimal. However, the long exposure times in actual repository systems preclude excluding radiolytic effects completely. Resolution of this issue will require accurate experimental data for the influence of pH on the kinetics of oxygen reduction on Alloy C-22.
- It has been shown that, in principle, the possibility that corrosion initiates and propagates on HLNW containers in the Yucca Mountain repository at short times (hundreds of years) when the temperature is significantly above the boiling temperature of water cannot be ignored. This is because surfaces are covered by highly hydrophilic oxides that will hydrate to form corresponding hydroxides, which are proton conductors and hence may act as electrolytes.
- A comparative experimental study of the electrochemical behaviors of Alloy C-22 and carbon steel has been carried out. The results show that Alloy C-22 has a unique resistance to pitting in NaCl solutions (concentration range to 1 mol/L, temperature range to 95 °C). Results thus far indicate that the corrosion rate of this alloy is characterized by a very low current density of alloy passive dissolution, which lies

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within the range of  $10^{-7}$  to  $10^{-8}$  A/cm. In neutral NaCl solutions, the reduction of dissolved oxygen is the most probable cathodic process. Basic kinetic parameters for anodic and cathodic processes have been determined.

### **Planned Activities**

The experimental measurements of parameters embodied in the point defect model (PDM) for Alloy C-22 and carbon steels will be performed during the second and third years of this project. The PDM postulates that passivity breakdown occurs as a result of cation vacancy condensation at the metal/oxide interface at sites in the passive film that are characterized by high cation vacancy fluxes. These sites correspond to regions of structural discontinuity.

Calculations on the velocities of pit and crack propagation as a function of the depth of penetration and environmental parameters will continue, along with the efforts on developing general damage function analysis theory.

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## A Single Material Approach to Reducing Nuclear Waste Volume

PI: James V. Beitz, Argonne National Laboratory

Project Start Date: August 1, 1999

Projected End Date: August 31, 2002

Project Number: 99-0219

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### Research Objective

This project is developing an innovative single material, minimum volume approach for the selective sorption of metal ion radionuclides from aqueous waste solutions and creation of a final nuclear waste form that is suitable for long term storage or geological burial. The project is based on a chemically functionalized porous silica that is termed Diphosil. Diphosil was created as an ion exchange medium that selectively and nearly irreversibly sorbs highly charged metal ions, such as actinides, from appreciably acidic aqueous solutions. The chelating power of Diphosil is due to diphosphonic acid groups that are anchored to its silica surface via organic spacer groups. Approximately 90 percent of the weight of dry Diphosil is silica ( $\text{SiO}_2$ ).

Underlying this project is the hypothesis that heating metal ion-loaded Diphosil in air will oxidize its organic content to water vapor and carbon dioxide and its phosphonic acid groups to phosphoric acid that would react with the sorbed metal ions to give metal phosphates. Based on literature reports of the properties of porous silica, it was further hypothesized that additional heating would either volatilize any excess phosphoric acid or cause it to react with the silica to form silicon phosphates. At still higher temperature, pore collapse should occur thereby microencapsulating and chemically fixing the sorbed metal ions in phosphate-rich metal phases in vitreous silica. Vitreous silica is one of the most radiation resistant glasses known.

### Research Progress

Project activities to date confirm the hypotheses as to the events that might occur when metal ion-loaded Diphosil is heated in air. The process of converting porous silica to fully dense silica is referred to as thermal densification in the literature because it occurs at temperatures far below the melting point of bulk silica and, in consequence, does not involve a phase change such as melting. The term thermal densification has been adopted to refer to the entire set of processes that occur when metal-ion-loaded Diphosil is heated in air to the point of pore collapse. Specific research conducted during the first year includes:

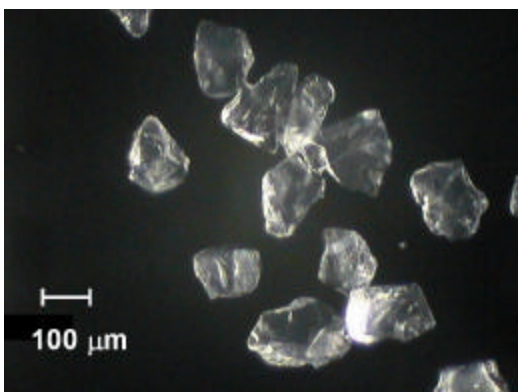
Solution Composition Effect on Metal Ion Sorption: This work has investigated the influence of solution composition variables on sorption of heavy metal ions into a chemically functionalized porous silica (Diphosil). Diphosil has been shown to extract



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metal ions from aqueous solutions that contain significant concentrations of ethylenediaminetetracetic acid (EDTA) at near-neutral pH. Aqueous solutions of EDTA are frequently used in decontaminating surfaces owing to its powerful chelating action for many metal ions. Using laser-induced fluorescence methods, evidence was obtained that Diphosil sorbs trivalent metal ions from concentrated phosphoric acid that contains a small concentration of nitric acid. This mixed acid media corresponds to the expected composition of the spent working medium of a nitric-phosphoric acid oxidation process for treating organic waste with significant plutonium contamination.

**Maximum Metal Ion Loading:** To determine the maximum heavy metal ion loading using Diphosil, optical spectroscopy was used to measure metal ion concentration during the sorption process. For example, the maximum uptake of trivalent neodymium ions ( $\text{Nd}^{3+}$ ) from dilute nitric acid was determined by monitoring a characteristic near-infrared optical adsorption band of  $\text{Nd}^{3+}$ .



**Photomicrograph of thermally densified, heavy metal ion-loaded Diphosil**

**Densification Optimization:** On-line, real time infrared analysis of the gases evolved during thermal densification of Diphosil in purified air as a function of heating rate and metal ion loading has been carried out. By varying heating and gas flow rates, optimal thermal densification conditions have been identified. The resulting material (see figure ) contains the selectively sorbed heavy metal ions in fully encapsulated nanophases that are embedded in nearly colorless, nonporous vitreous silica that is highly resistant to radiation damage.

### Planned Activities

Diphosil possesses the unusual ability to selectively and strongly sorb high valent metal ions, such as actinides, from aqueous solutions. Assessments of the full range of aqueous waste solutions that are amenable to processing via Diphosil are being pursued. During the second year, research will be completed on the influence of monovalent (1+) and divalent (2+) metal ions on the uptake of more highly charged metal ions by Diphosil in aqueous solutions. This work will emphasize common alkali and alkaline earth ions, such as  $\text{Na}^+$  and  $\text{Ca}^{2+}$ , over a range of pH values that extend from significantly acidic to nearly neutral.

Densification studies will be completed by determining the temperature and time at temperature required to achieve complete densification. Samples containing high specific activity, short-lived radionuclides will be prepared as the starting point for radiation damage studies on thermally densified, metal ion-loaded Diphosil. As these short-lived radionuclides decay, it is anticipated that laser-based high resolution linear and nonlinear optical studies will quantify the effect of the resulting radiation damage on

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the local environment of the encapsulated heavy metal ions. To better understand the observed complex luminescence decays from the encapsulated metal ions, a theoretical modeling effort will begin on the basis of dipole-dipole energy transfer. Conventional leach rate studies will quantify retention of metal ions within thermally densified Diphosil prior to and following damage due to ionizing radiation.